University of Pannonia
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Magnetic Colloids in External Electromagnetic Fields

Ph.D. THESIS

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Introduction

The term of intelligent, adaptive or clever material was first used in the 1980s to refer to materials that adapt to environmental changes by altering their properties. Soft smart materials belong to the large group of smart materials; they can be utilized in several ways. One of the special types of intelligent materials are the ferrofluids, also known as magnetic fluids. Ferrofluids are colloidal liquids made of nanoscale magnetic particles suspended in a carrier fluid, forming a stable system. The particles have a monodomain structure and therefore exhibit superparamagnetic properties. Nowadays, there is high demand in industrial and medical applications for the development of the above mentioned materials. The main directions of the research of these are focusing on the production possibilities, characterization of materials and the exploration of the areas of use. This research is focused on intelligent materials that are sensitive to magnetic fields. These materials are able to change their macroscopic parameters under the influence of a magnetic field.

Aims

As different properties are used in each application, the aim of the research was to determine the magnetic field-dependent properties of the most important parameters of magnetic fluids. During the experimental work, three major goals were set. The force acting on the magnetic fluid droplets in an inhomogeneous magnetic field will be determined. The change in the contact angle of the magnetic liquid droplets due to the magnetic field will be measured on different surfaces. Another goal was to develop and test a measurement system for the study of magnetic liquids for hyperthermia.

Experiments

The experimental work entails three major areas, similar to the aims. In the first part, the force acting on a magnetic fluid droplet was determined by using an inhomogeneous magnetic field, a property that can be important in biomedical applications. During the experiments, the force acting on magnetic droplets of different sizes at different field strength values was measured, and then the experimental data was compared with theoretical calculations. The aim was to develop a model that can be used to estimate the interactions between the droplet and its environment by knowing the properties of the liquid droplets.

In the framework of the second topic, the change of the contact angle of magnetic liquid droplets was measured under the influence of magnetic field on hydrophilic and on hydrophobic surfaces. The purpose was to determine the contact angle and by this the field-dependence of the wetting properties.

The third topic centers on the development of a measurement system using differential cell and differential temperature measurement for the therapeutic hyperthermia study of magnetic fluids. The objective of the research was to develop a low-cost method that can be used in an alternating magnetic field, which allows similar or better accuracy to the currently used temperature measurement methods.

New scientific results, theses

1. Using a self-developed device, the force acting on biocompatible magnetic fluid droplets of different sizes at several field strength values was measured. (Relevant publications: **[C3]**, **[K5]**)

1.1. It was found that the force acting on ferrofluid droplets can be approximated with acceptable accuracy by the Langevin magnetization of the magnetite particles calculated with appropriate parameters, if the field strength is calculated by the modified mean field (MMF2) model.

In low intensity non-uniform magnetic field with (39.9-83.1) kA m^{-1} field strength and (810-7195) kA m^{-2} gradient, the tendency of the theoretical and experimental data was almost the same (within 10%). In medium intensity non-uniform magnetic field with (79.4-169.3) kA m^{-1} field strength and (1734-15027) kA m^{-2} gradient, close to the pole of the magnet at high gradient and above 150 kA m^{-1} field strength the measured values were greater than the calculated data, moving away from the pole of the magnet at low gradient and below 100 kA m^{-1} field strength the measured values were smaller than the calculated values. In high intensity non-uniform magnetic field with (117.7.9-251.0) kA m^{-1} field strength and (2440-21929) kA m^{-2} gradient, close to the pole of the magnet at high gradient and above 175 kA m^{-1} field strength the measured values in the case of the 5.19 m/m% concentration sample were higher compared to the theoretical calculation, but moving away from the pole of the magnet at low gradient and below 150 kA m^{-1} field strength the course of the calculated and measured data is almost the same (within 10%).

1.2. During the measurements, three types of magnetite concentrations were examined with three different droplet sizes. It was found that at the highest droplet size (73 μl), the model overestimates the measured values at low concentrations (2.58 m/m%) and underestimates at higher concentration (5.19 m/m%).

2. Using self-developed devices, the change of the contact angle of magnetic liquid droplets was measured under the influence of magnetic field on hydrophilic (silicon single crystal wafer) and on hydrophobic (PDMS) surfaces at different temperatures. (Relevant publications: **[C2]**, **[K2]**, **[K3]**, **[K4]**)

2.1. At low field strength (up to 10 kA m^{-1}), it was found that the contact angles decrease in proportion to the magnetite concentration with increasing field strength for both surface types. In the case of PDMS surface, there is a difference in the tendency of the measured values compared to the silicon single crystal surface at the highest colloid concentration (5.80 V/V%).

2.2. In the case of high field strength (above 10 kA m^{-1}), the tendency of the change of the contact angle on the hydrophobic surface was reversed (there is a local minimum). In the highest examined concentration (5.80 V/V%), contact angles larger than the initial ones were also measured. On hydrophilic surface, this phenomenon was observed in case of the undiluted sample. The tendency of the change of contact angle depends on the temperature on both surfaces, however, the local minimum appears at different field strength values in the case of different surfaces. The minimum appears at

(35-40) kA m^{-1} field strength in the case of silicon wafer. The minimum appears at lower field strengths in the case of PDSM surface: between (10-20) kA m^{-1} field strength at 15°C and between (5-10) kA m^{-1} field strength at 24°C.

3. The SLP values of organic solvent and water-based ferrofluids were determined by using a differential cell instrument and a self-developed differential temperature measurement method. The two-coil arrangement allows the reference material to be measured at the same time as the colloid, thus eliminating the thermal effect affecting the measurement result. (Relevant publications: **[C1]**, **[K1]**)

3.1. It was found that differential cell arrangement and differential temperature measurement could be effectively applied to measurement of dilute colloids (5.80 V/V%) and measurements in low intensity magnetic fields (0.32 kA m^{-1}). Measurements were performed at frequency (480±1 kHz) and field strength values appropriate to clinical conditions.

3.2. It was found that temperature sensors (T and K type thermocouples and Pt100 RTD, which are normally sensitive to high-frequency fields due to their material) in differential mode could also be used to measure relative temperatures in an alternating magnetic field (300 kHz-800 kHz). During the experiments, it was found that the alternating magnetic field can significantly affect the measurement results when using individual temperature sensors.

3.3. The self-heating caused by the alternating magnetic field can be compensated in the differential configuration, thus, it is suitable for monitoring the relative temperature change without other additional measurements.

3.4. The SLP values (between 0.050 W g^{-1} and 0.093 W g^{-1}), which are calculated from the temperature change measured with different temperature sensors, showed a good agreement. It was found that the SLP values were inversely proportional to the concentration of the colloids: there is an increase of 77% and 57% in the diluted samples in the case of EMG700 and EMG900 colloids, respectively. The ILP values are consistent with the literature.

3.5. The applicability of the differential measurement method at higher field strengths was verified by measuring the colloid EMG900-100 at (0.53±0.02) kA m^{-1} field strength at a frequency of 480 kHz. The SLP value of the colloid was (0.131±0.004) W g^{-1}. Compared to the data measured at 0.32 kA m^{-1} field strength, a quadratic dependence of the SLP on the magnetic field strength was observed.

Scientific publications, presentations and posters for the basis of theses

[C1] **Guba, S.**; Horváth, B.; Molnár, G. and Szalai, I.: A double cell differential thermometric system for specific loss power measurements in magnetic hyperthermia, Measurement, (2020), IF (2019): 3,364, Q1
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[C2] **Guba, S.**; Horváth, B. and Szalai, I.: Examination of contact angles of magnetic fluid droplets on different surfaces in uniform magnetic field, Journal of Magnetism and Magnetic Materials, (2019), IF (2019): 2,954, Q2
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[C3] **Guba, S.**; Horváth, B. and Szalai, I.: Determination of the Force Acting on Biocompatible Ferrofluid Droplets in Inhomogeneous Magnetic Field, Journal of Magnetism and Magnetic Materials, 444 (2017), IF (2017): 3,233, Q1
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[K1] **Guba, S.**; Horváth, B.; Molnár, G. és Szalai, I.: Mágneses folyadékok melegedésének vizsgálata váltakozó mágneses térben, XXI. Energetika-Elektrotechnika – ENELKO és XXX. Számítástechnika és Oktatás – SzámOkt Multi-konferencia

[K2] **Guba, S.**; Horváth, B. és Szalai, I.: Mágneses folyadékcseppek kontaktszögének meghatározása mágneses térben különböző hőmérsékleteken In: Dr. Sebestyén-Pál, György; Dr. Szabó, Loránd; Dr. Biró, Károly-Ágoston (szerk.) ENELKO 2019 SzámOkt 2019: XX. Nemzetközi Energetika-Elektrotechnika Konferencia, XXIX. Nemzetközi Számítástechnika és Oktatás Konferencia, Erdélyi Magyar Műszaki Tudományos Társaság (EMT), (2019) pp. 38-44., 7 p., Kolozsvár, Románia, 2019. október 10-13.

[K3] **Guba, S.**; Horváth, B. and Szalai, I.: Examination os contact angles of magnetic fluid droplets on different surfaces in uniform magnetic field, International Conference on Magnetic Fluids – ICMF 2019, Párizs, Franciaország, 2019. július 8-12.

[K4] **Guba, S.**; Horváth, B. és Szalai, I.: Mágneses folyadékcseppek kontaktszögének meghatározása különböző felületeken In: Barabás, István (szerk.) XXVII. Nemzetközi Gépészeti Konferencia OGÉT 2019,: Erdélyi Magyar Tudományos Társaság (EMT), (2019) pp. 153-156., 4 p. Nagyvárad, Románia, 2019. április 25-28.

[K5] **Guba, S.**; Horváth, B. és Szalai, I.: Biokompatibilis szuperparamágneses folyadékcseppekre ható erő meghatározása inhomogén mágneses térben, In: Biró, K Á; Sebestyén, P Gy (szerk.) ENELKO 2016 - XVII. Nemzetközi Energetika-Elektrotechnika Konferencia, SzámOkt 2016 - XXVI. Nemzetközi Számítástechnika és Oktatás Konferencia, Kolozsvár, Románia: Erdélyi Magyar Műszaki Tudományos Társaság (EMT), (2016) pp. 47-52., 6 p., ENELKO, Kolozsvár, 2016. október 6-9.

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2. Méndez-Bermúdez, J. G., Dominguez, H.; Pusztai, L.; **Guba, S.;** Horváth, B. and Szalai, I.: Composition and temperature dependence of the dielectric constant of 1-propanol/water mixtures: Experiment and molecular dynamics simulations, Journal of Molecular Liquids 219 pp. 354-358, (2016) http://dx.doi.org/10.1016/j.molliq.2016.02.053
3. Horváth, B.; **Guba, S.** és Szalai, I.: Mágneses folyadékok és szuszpenziók viselkedése külső mágneses térben, Balogh, András; Klein, Mónika (szerk.) Konferencia Kiadvány (2019) p. 43, Műszaki Kémiai Napok 2019, Veszprém, 2019. április 16-18.
4. Horváth, B.; **Guba, S.**; Rigó, M. és Szalai, I.: Mágneses folyadékok és szuszpenziók mágneses és reológiai tulajdonságai, Műszaki Kémiai Napok, Veszprém, 2018. április 24-26.
5. **Guba, S.**; Horváth, B. és Szalai, I.: Mágneses részecskéket tartalmazó kompozit elasztomerek készítése és mechanikai vizsgálata, XXIII. Nemzetközi Vegyészkonferencia, Déva, Románia, 2017. október 25-28.
6. **Guba, S.**; Horváth, B. és Szalai, I.: N-ZVI tartalmú kompozit elasztomerek vizsgálata, Energetika-Elektrotechnika Konferencia - ENELKO 2017, Nagyvárad, 2017. október 12-15.
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8. **Guba, S.**; Horváth, B. és Szalai, I.: Biokompatibilis szuperparamágneses folyadékcseppekre ható erő meghatározása inhomogén mágneses térben, PhD hallgatók anyagtudományi napja XVI., Veszprém, 2016. november 28.